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Abstract

A study of JA2 ignition using high-power (3 kW) pulsed Nd:glass lasers at low loading densities was undertaken to help construct models for predicting initial pressurization in bombs at high loading densities. Comparisons were made with results of experiments using a low-power (0.1 kW) continuous wave (cw) CO_2 laser. Differences in wavelength dependence of optical properties and time dependence of the laser pulse were considered. Measured infrared (IR) emission delays agreed with the calculated "reaction" delays using a condensed-phase model. The calculations are sensitive to values used for optical properties. Experiments in which laser energy and beam diameter were varied indicate that there is an upper limit on flux density values for flamespreading to occur. Exceeding the limit leads only to ablation. An understanding of the ablation process due to high-power irradiation of energetic solids is needed to predict the pressure response of solid propellant to high-power lasers.

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1. Introduction

The use of laser radiation is being considered as a replacement for primer materials in some igniter trains [1]. This is expected to lead to better performance and further increase in safety. The possibility of multipoint ignition and the ability to control ignition timing may offer the interior ballisticians a variety of methods for improving performance. Direct radiant ignition of several nitrate-ester propellants with a pulsed Nd:glass laser has been reported [2]. The experiments were done in small closed bombs (1–10 cm^3) at high loading densities (1 g/cm^3) and low porosities (0.4). The propellant surfaces were graphite coated to increase radiant absorption. For a given pulse duration and laser beam diameter, reliable ignition required laser energies that generated pressures in the bombs greater than 10 MPa prior to deradiation. Radiant ignition models that are able to predict the initial pressurization in closed bombs may be helpful in scaling these results to larger gun chamber volumes.

This report attempts to relate recent results of ignition experiments with JA2, a nitrate ester propellant, using high-power (3 kW) pulsed Nd:glass lasers with results of experiments using a low-power (0.1 kW) continuous wave (cw) CO_2 laser. Differences in wavelength dependence of optical properties and time dependence of the laser pulse are considered. Surface temperatures calculated from measured emission delays using the CO_2 laser are compared with those calculated for the Nd:glass laser experiments. Measured infrared (IR) emission delays are compared with predictions of a phenomenological condensed-phase radiant "ignition" model [3, 4]. The model is described in Vilyunov and Zarko [3] It considers condensed-phase (surface) reactions and in-depth energy absorption but neglects energy losses (i.e., those losses due to surface [film] heat transfer and emission) and propellant consumption and does not consider the effect of laser interruption (deradiation). The model is one-dimensional (1-D) (semi-infinite solid). It assumes that flux is continuous and uniform and that energy production can be described by a first-order (global) surface reaction. The relevant energy equation and boundary conditions used for (transparent) solids are:

$$c\frac{\partial T}{\partial t} = \alpha c\frac{\partial^2 T}{\partial x^2} - \frac{1}{\rho}\frac{\partial q}{\partial x} + Qz\exp\left(-E/RT\right)$$

and

$$T(x,0) = T_o;\ \frac{\partial T}{\partial x}(0,t) = 0;\ \ T(\infty,t) = T_o,$$

where $q = q_o \exp(-nx)$, q_o = transmitted surface flux, n = absorption coefficient, T = temperature, t = time, x = distance from irradiated surface, T_o = ambient temperature, α = thermal diffusivity, ρ = density, and c = specific heat. The last term in the energy equation is the Arrhenius global heat release rate during ignition (E = activation energy, z = first-order pre-exponential factor, and Q = heat of reaction).

The ignition delay (t_{ig}) is the sum of (inert) "heat up" (t_h)and chemical induction (t_{ch}) times. The surface temperature (T_h)and t_h are determined by finding the temperature at which the time derivatives obtained from analytic solution of the energy equations corresponding to inert (radiant) heating ($Q = 0$) and to adiabatic (spatially independent) reaction ($\alpha = q_o = 0$) become equal. T_h represents the chemical reaction temperature at which the (specific) rate of chemical energy production equals the radiant energy absorption (at the surface), and t_h is the time required to reach this temperature. At later times, due to the Ahrrenius exponential temperature dependence, chemical rates dominate and it is assumed that surface reaction occurs under adiabatic conditions. The chemical induction time is obtained from the adiabatic solution and is defined (in this model) as the time (with respect to t_h) for time derivative to become infinite. Approximations to these analytic solutions (given in Vilyunov and Zarko [3]) were used to calculate T_h and the corresponding t_h and t_{ig} as a function of radiant flux. The inert approximation (for T_s and t_h) is valid under conditions when characteristic lengths for heat conduction during heat up are much greater than those for absorption $\left(n\sqrt{\alpha t_h} >> 1\right)$. The adiabatic approximation (for t_{ch}) is valid when activation energies are much greater than thermal energies $\left(E/RT_h >> 1\right)$. The model (often referred to as "adiabatic") was used with some success to predict the dependence of M9 emisson delays measured in a closed bomb (at low loading densities) on radiant flux density from the CO_2 laser [4].

2. Experimental Technique

JA2 (0.60 nitrocellulose [NC]/0.15 nitroglycerine [NG]/0.25 diethylene glycol dinitrate [DEGDN]) and inert (polymer without oxidizer) propellant samples (1/8 in wide) were cut from solid cylinders (1/4 in diameter). The flat surface was mounted perpendicular to the laser beam. Two nominally 30-J lasers were used with pulse durations of 5 ms (LRS-90, HiShear Technology Corp.) and 10 ms (GNL-10, Laser Photonics, Inc.). A focusing lens was used to vary the flux intensity by changing beam diameter at the sample location. A silicon diode (J14, EG&G Judson), which is sensitive to radiation between 0.5 μm and 1.1 μm, was used to record the temporal (relative) intensity profile by measuring reflected radiation from the lens. An energy meter (Scientech) was used to correlate output energy with voltage settings. Experiments were done in ambient air or in a 1-liter closed bomb in air, N_2, or Ar at 0.1 MPa. The bomb, CO_2 laser and the measurements of emission (0.3-0.8 μm) and pressure have been previously described [4]. In addition, a photoconductive (HgCdTe) detector (J15D12, EG&G Judson) was used with an interference filter to monitor IR emission centered at 4.49 μm. This wavelength region contains strong emission bands of the expected combustion products CO and CO_2

3. Results

3.1 Model Calculations/CO_2 Laser Experiments. Figure 1 shows the comparison of the inert reaction delays calculated from the adiabatic model with JA2 emission delays obtained from experiments with the CO_2 laser. Also shown are the effects of values for the Ahrrenius rate constant and the absorption coefficient on calculated results. The experiments were done in the bomb using air and Ar at initial pressures of 0.1 MPa. The calculations used generally accepted values for thermophysical properties (heat capacity = 0.33 cal/g-K, thermal conductivity = 0.0005 cal/cm-s-K and thermal diffusivity = 0.001 cm^2/s; measured values at 10.6 μm [4] for optical properties (absorption coefficient [n] = 250 cm^{-1} and reflectance [R] = 0.08); and Ahrrenius kinetic parameters, determined previously from shock-tube experiments with NC for the global energy production rate [5]. The calculation for n = 15 cm^{-1}, which is the experimentally derived value at 1.06 μm [6], is

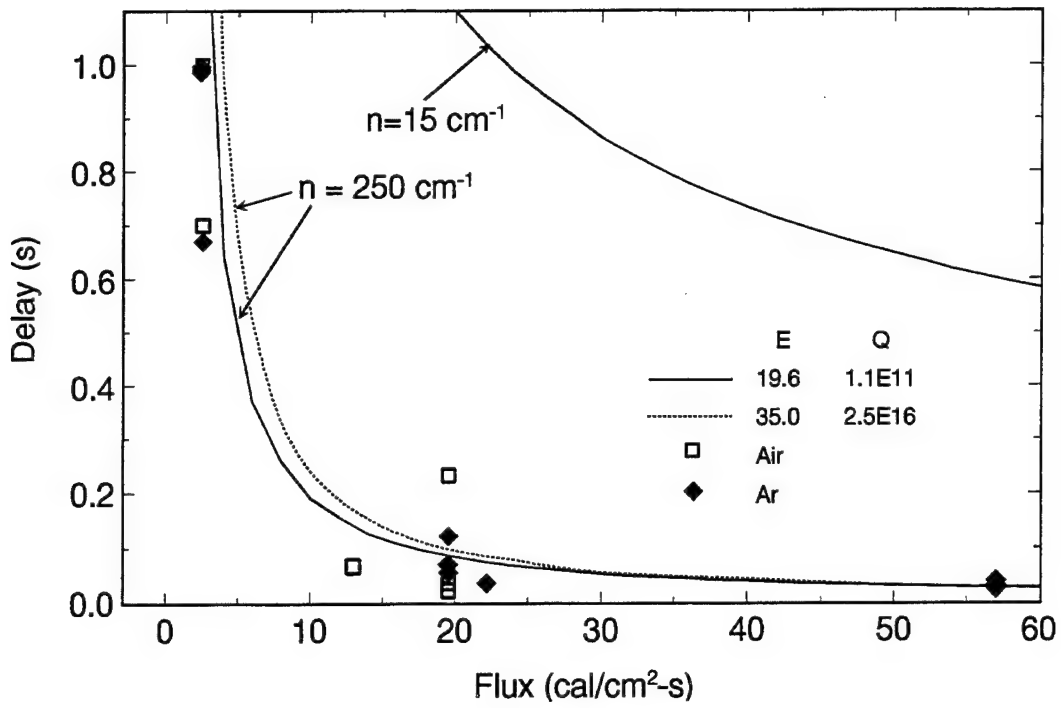

Figure 1. Comparison of Adiabatic Model Calculations and Measured JA2 Emission Delays.

presented to show the effect of n on the calculations. The calculation using kinetics derived from radiant ignition experiments with N propellant (a former Soviet Union [FSU] triple base similar in composition to JA2) is presented to show the effect of kinetic parameters (E = activation energy [kilocalorie/mole], Q = pre-exponential factor [calorie/gram-second]) on the calculations. In addition to the inert reaction delay, an ignition delay can also be calculated. The difference between them is equal to the adiabatic explosion delay (from Thermal Explosion Theory [3]). For global kinetics associated with energetic materials, this is less than 25% of the ignition delay.

The surface temperatures at the time of the measured emission delays in air (shown in Figure 1) were calculated, assuming the grains were inert. Figure 2 shows the flux density dependence of these calculated surface temperatures. The lines are the average temperatures of the data and a linear fit to the data, which can be considered as a constant or a flux-dependent critical temperature (T_{crit}) criterion for initiation of emission/reaction, respectively. Figure 3 compares calculated delays based on the surface temperature given by the linear fit to the air data with Ar (and air) data. Also shown

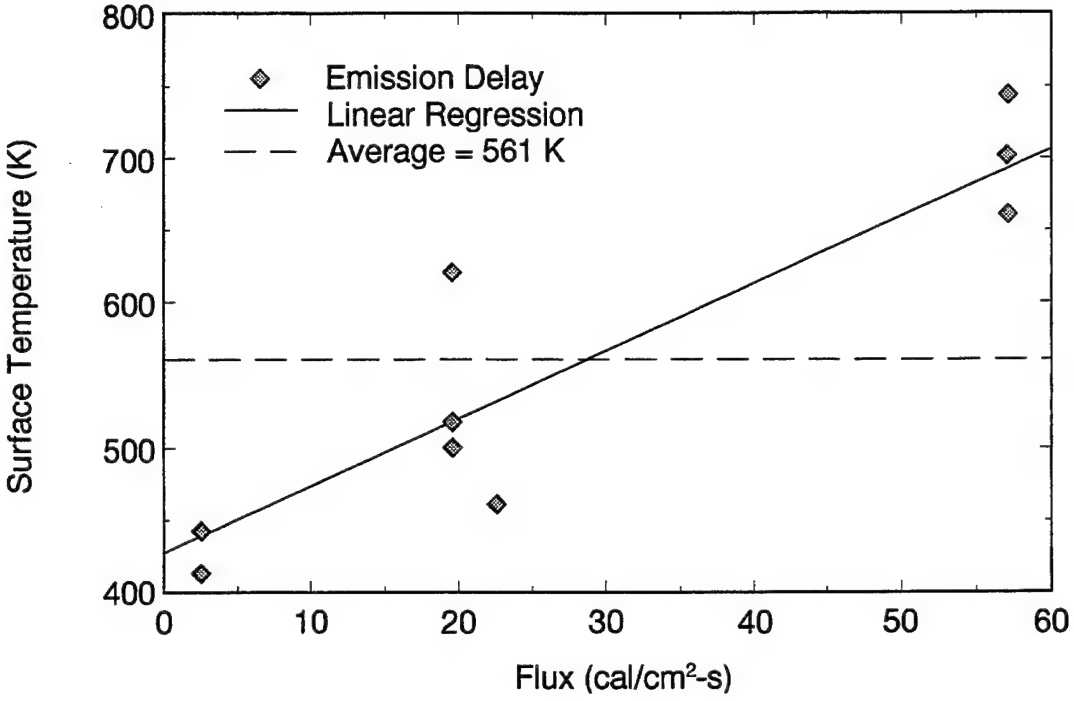

Figure 2. Dependence of the Calculated Surface Temperatures Corresponding to Measured Emission Delays (in Air) on the CO_2 Laser Flux Density.

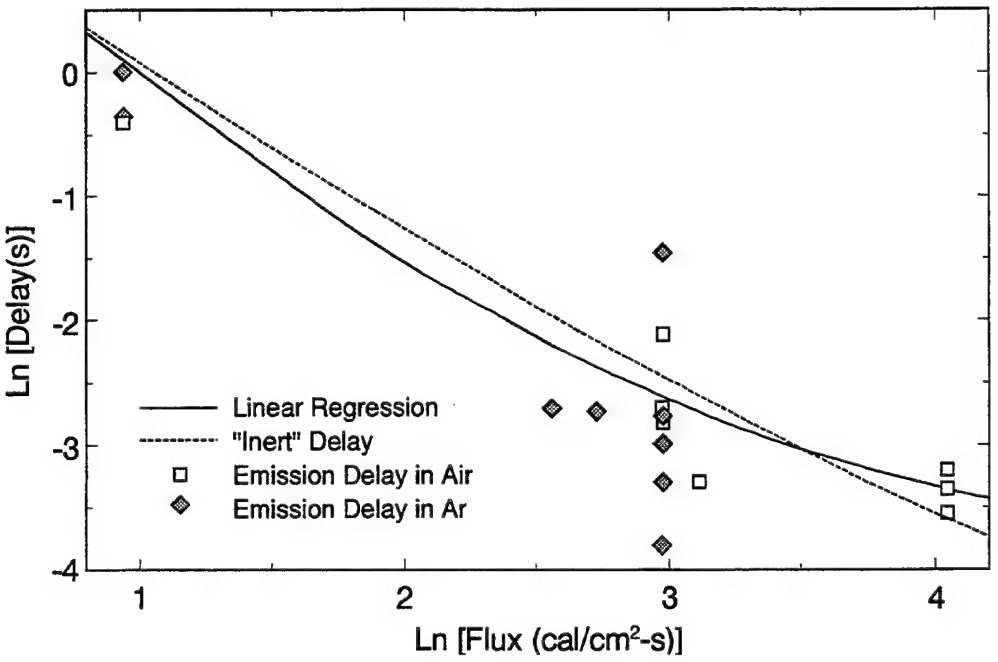

Figure 3. Comparison of Flux Density Dependence of Measured Emission Delays in Air and Ar With Critical Temperature (Air Data) and With Model Predictions.

are the inert delays calculated from the adiabatic model. All of the calculations (delays and surface temperatures) make use of the analytic solution for the transient response of a semi-infinite, semi-transparent body under constant, uniform radiative heating (a 1-D problem) [7].

The calculation of ignition delays (adiabatic model) for the Nd:glass data requires the corresponding two-dimensional (2-D) solution for a pulsed beam with Gaussian spatial distribution. It appears that the solution for the semi-transparent (in-depth absorption) problem is not known. The solution for the opaque problem is given by Ready [8].

The delays and surface temperatures for in-depth absorption with Gaussian beams have been calculated, assuming ignition occurs at the beam axis and employing a correction factor for the Ready solution (opaque). The factor is the ratio of surface temperatures calculated for semi-transparent and opaque bodies obtained from the analytic solution for the uniform spatial distribution problem given by Boehringer [7]. This ration depends on the absorption coefficient and is independent of the flux density. Figure 4 shows the ratios calculated from the 1-D solution for $n = 5$, 15, and 50 cm^{-1}.

The spatial energy distribution of the GNL-10 laser (focused) beam was measured at the sample location in the bomb. It gave a reasonable fit to a Gaussian distribution. The derived values for the Gaussian beam radius $\left(g = FWHM / 2\sqrt{\ln(2)}\right)$ and the axis energy (E_o) (at maximum capacitor voltage) were 3.2 mm and 13.5 J. The radiant-axis flux density (F_o) was obtained by dividing E_o by the pulse duration (11 ms) and the area of the aperture (4.2 mm) that was used to determine the spatial energy distribution.

Figure 5 shows the temporal intensity of the GNL-10 laser as recorded by the silicon diode. The maximum voltage and pulse duration (11 ms) have been used to obtain nondimensional units. The intensity fluctuations are probably due to a voltage supply problem, but they do not change over time. The smooth curve is a polynomial fit to intensity. This fit was used to calculate the (nondimensional) time dependence of surface temperature at the axis of a Gaussian beam for opaque bodies. The reference temperatures and times depend on the Gaussian parameters F_o and g. The

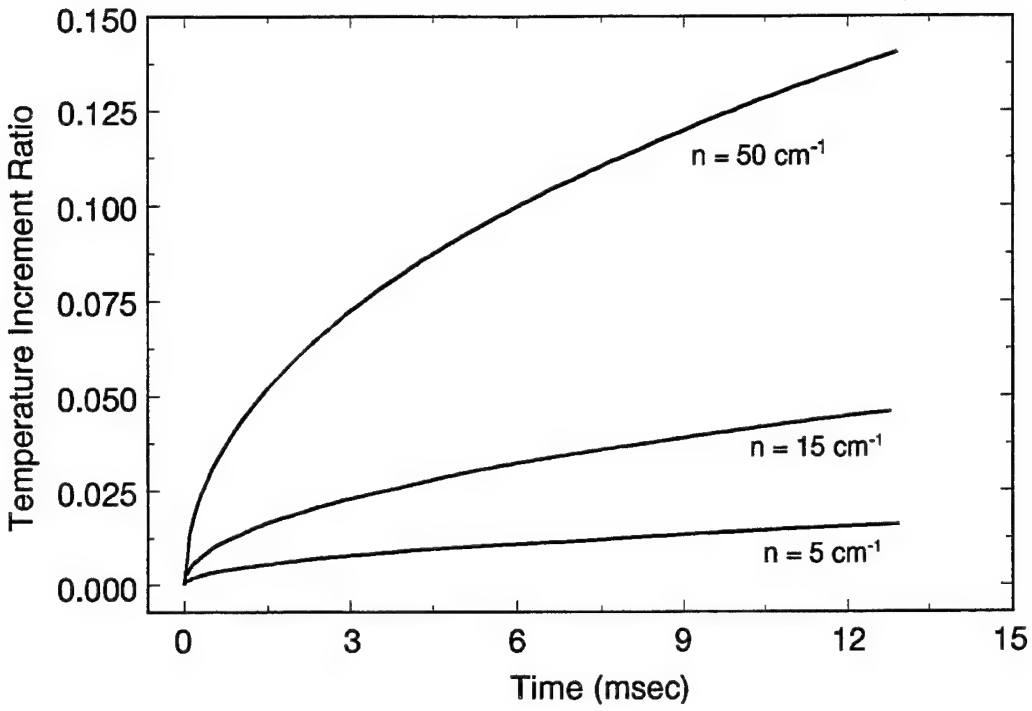

Figure 4. Effect of Absorption Coefficient on the Ratio of the Semitransparent-to-Opaque Propellant Surface Temperature for Constant, Uniform Irradiation.

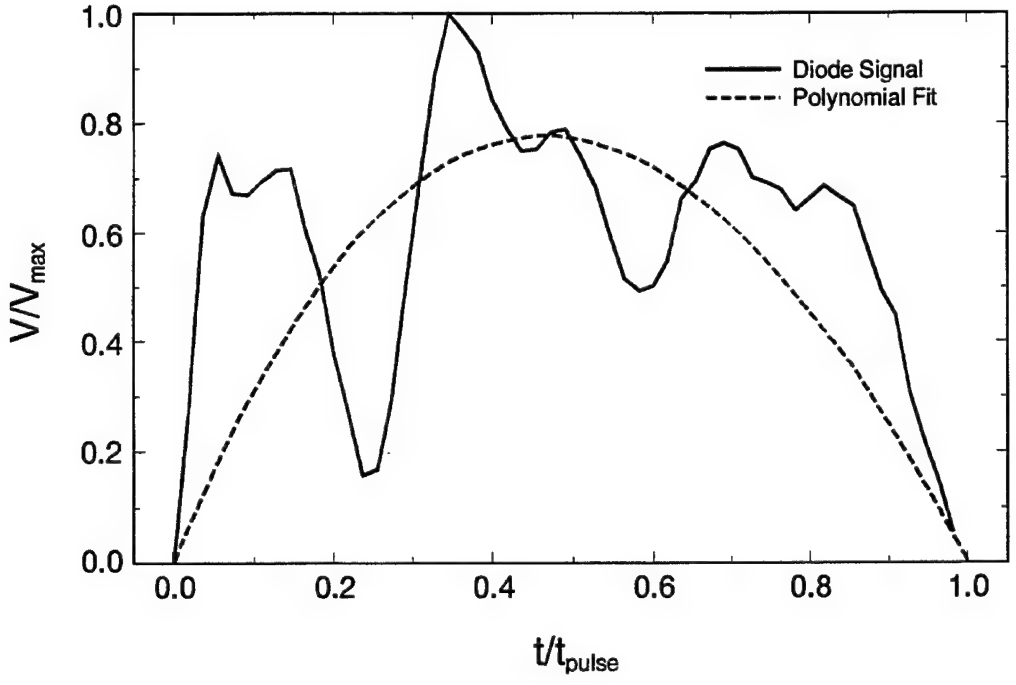

Figure 5. Silicon-Diode Signal and Polynomial Fit of Radiation Intensity During a 10-ms Nd:Glass Laser Pulse.

effect of in-depth absorption (n) was obtained by multiplying the transient surface temperature for opaque bodies by the corresponding ratio shown in Figure 4.

Figure 6 shows the effect of n on the calculated surface temperature at the beam axis (T_b). The dotted line is T_{crit}, the average "reaction" critical surface temperature (in Figure 2).

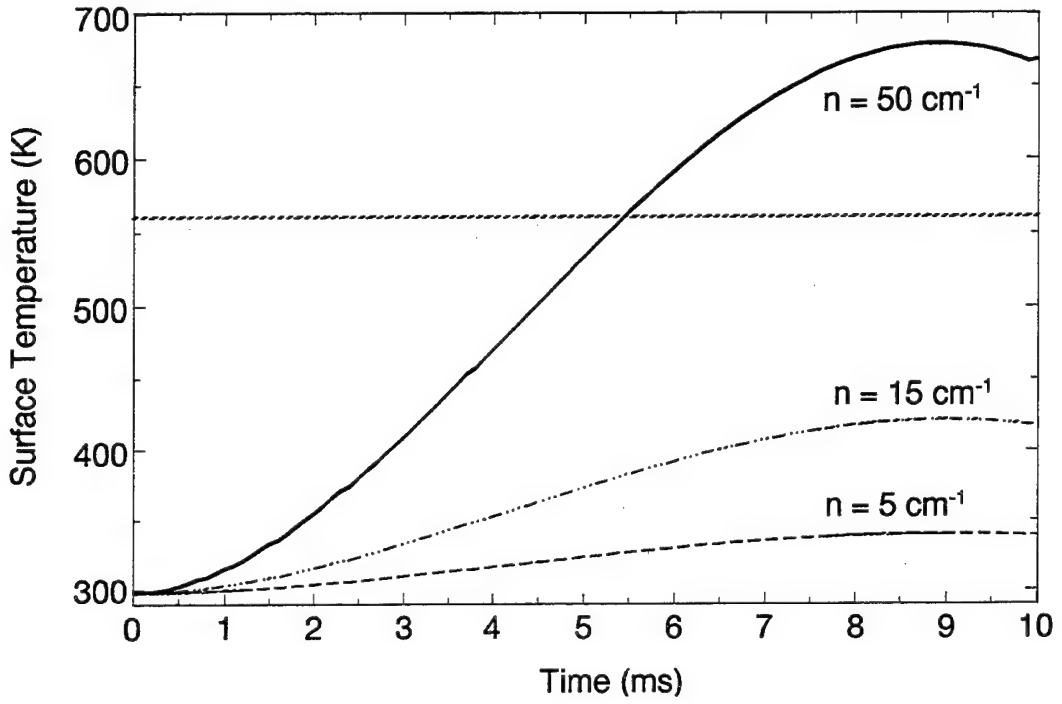

Figure 6. Effect of Absorption Coefficient on Calculated Inert Surface Temperature at the Axis of the Pulsed Gaussian Laser Beam; Energy = 13.5 J, Beam Diameter = 3.2 mm.

The inert (reaction) delays (t_{in}) based on the adiabatic model were calculated. The model requires that t_{in} be less than the time at which the maximum in T_b occurs (t_{max}), which is 9 ms. These delays correspond to the T_b value at which the time derivatives of T_b (obtained from polynomial fits to curves in Figure 6) and time derivative of the adiabatic reaction temperature are equal. The latter derivative is given by the product of (Qz/c) and exp $[-E/R(T_b)]$. The dependence of these delays on F is shown in Figure 7. The apparent falloff in the flux dependence of t_{in} at low flux values is probably due to difficulties of obtaining meaningful derivatives from the polynomial fits near maxima (t_{in} = 9 ms).

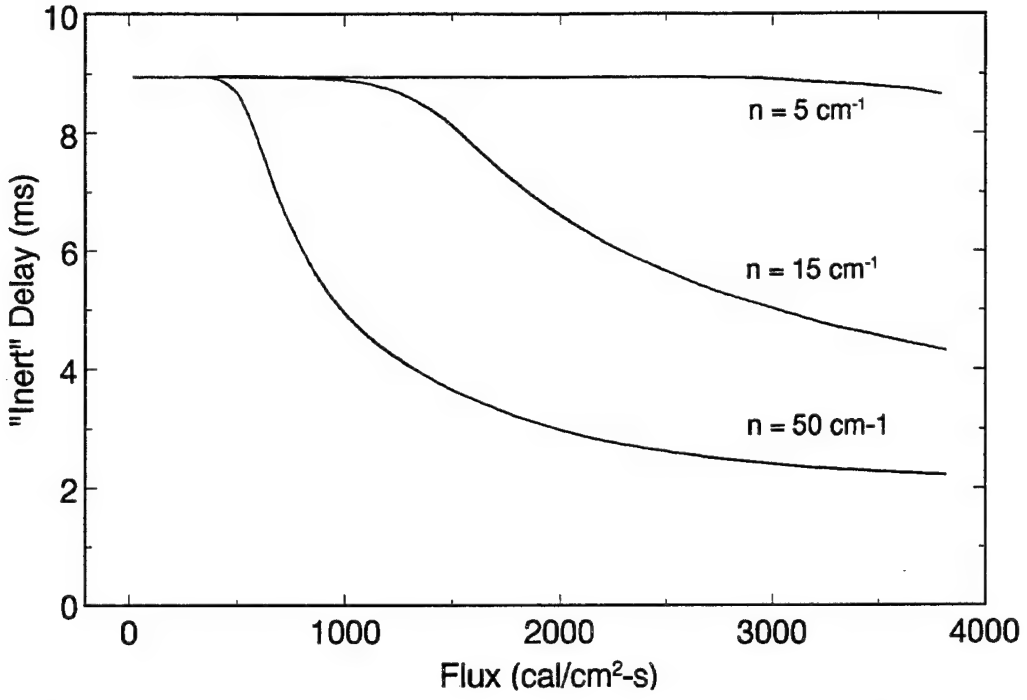

Figure 7. Flux Dependence of the Adiabatic Model Inert (Reaction) Delay Calculated for Transient Surface Temperatures in Figure 6.

3.2 Nd:Glass Laser Experiments. Figure 8 shows pressure signals from experiments in the bomb with JA2 and an inert (thermophysical properties similar to energetic propellants) propellant with air and N_2 at an initial gas pressure of 0.1 MPa. A low-pass digital filter was used to minimize the effects of noise in the pressure signal generated by capacitor discharge during the laser pulse. It is believed that filtering affects measurements of pressure delays (initial rise time) more than the initial pressurization rates. The lower flux densities were obtained with the GNL-10 laser (10-ms pulse) and the higher one with the LRS-90 laser (5-ms pulse).

Figure 9 shows the corresponding photomutiplier signals for the 10-ms pulse experiments ($F = 2.7$ kcal/cm^2-s) in Figure 8. Only the baseline region is shown, and the emission fluctuations, which follow the laser intensity fluctuations, are omitted.

Figure 10 shows the corresponding photoconductive detector signals for the 10-ms pulse experiments in Figure 9. The silicon-diode signal for the JA2/N_2 experiment is also shown. The amplitude of each signal shown is relative to its value at 1 ms.

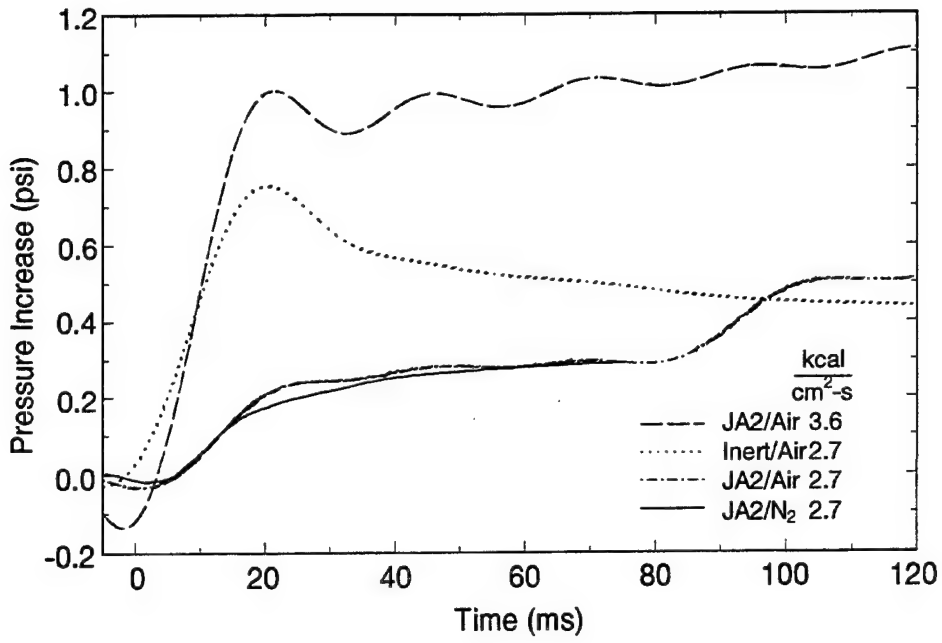

Figure 8. Pressure Signals From Experiments in the Closed Vessel That Show Differences in the Response of JA2 and an Inert Propellant to a 10-ms Laser Pulse.

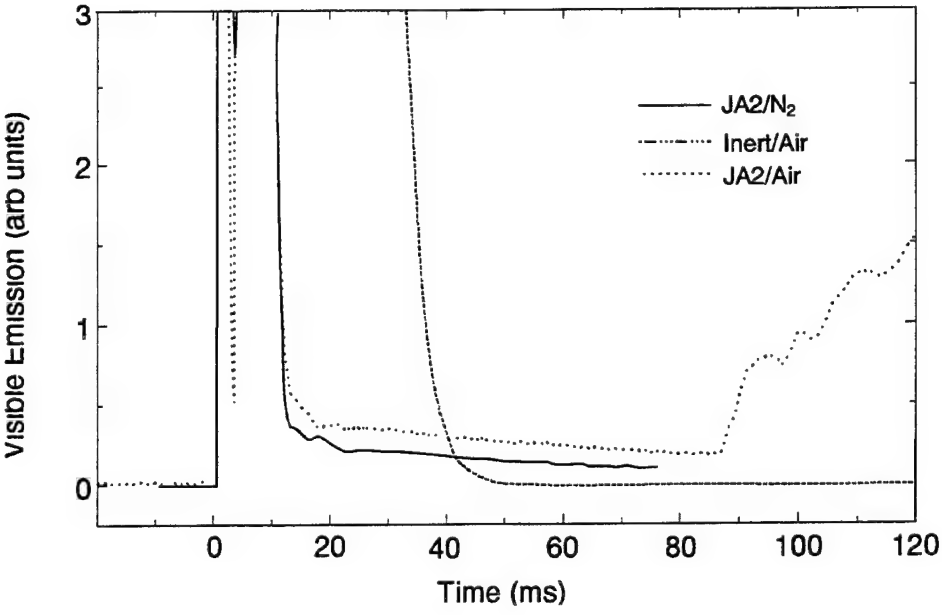

Figure 9. Visible Emission From JA2 and an Inert Propellant in the Closed Vessel During a 10-ms Laser Pulse.

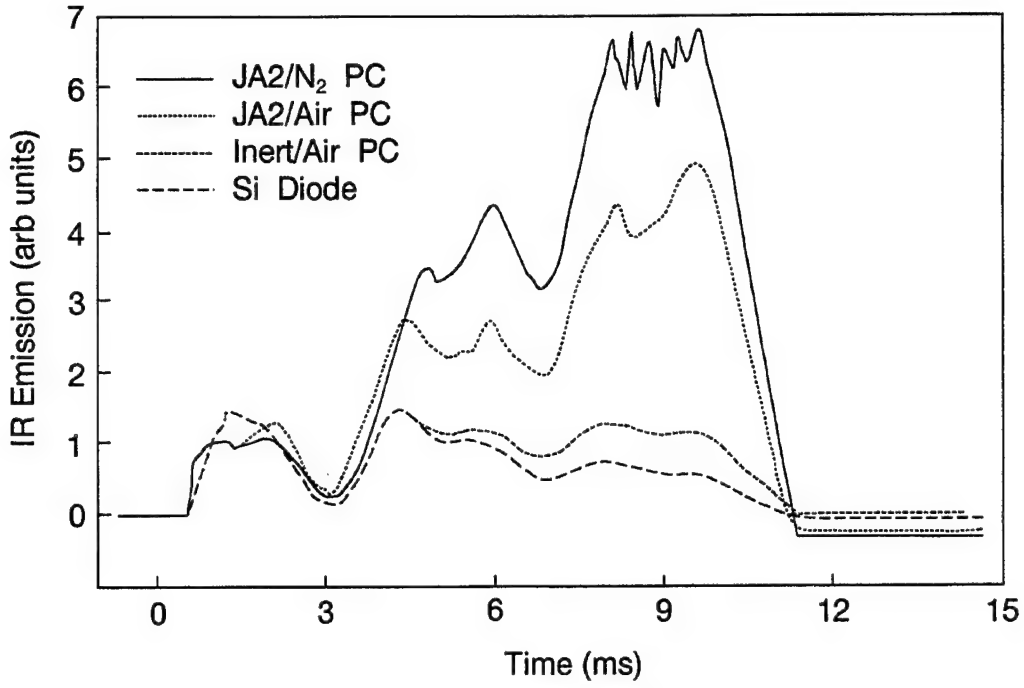

Figure 10. IR Emission at 4.49 μm From JA2 and an Inert Propellant During a 10-ms Laser Pulse in the Closed Vessel. Intensities Are Relative to Values at 1 ms. Note the Increase in JA2 Emission Near 4 ms, Which Is Attributed to Initiation of Exothermic Reactions.

Figure 11 shows photoconductive detector signals for 10-ms pulse experiments ($F = 2.0$ kcal/cm^2-s) with JA2 and an aluminum disk in room air. The silicon-diode signal (proportional to the laser intensity) for the JA2 experiment is also shown. The amplitude of each signal shown is relative to its value at 1 ms.

Flux values (F) for initiation of flamespreading with the GNL-10 laser were limited to less than 5,000 cal/cm^2-s. This is due, in part, to energy limitations of the laser. Increasing F (at fixed energy) by decreasing the beam radius (g) does not necessarily increase the probability for flamespreading. This can be seen from the results of experiments with JA2 in ambient (room) air, which are listed in Table 1.

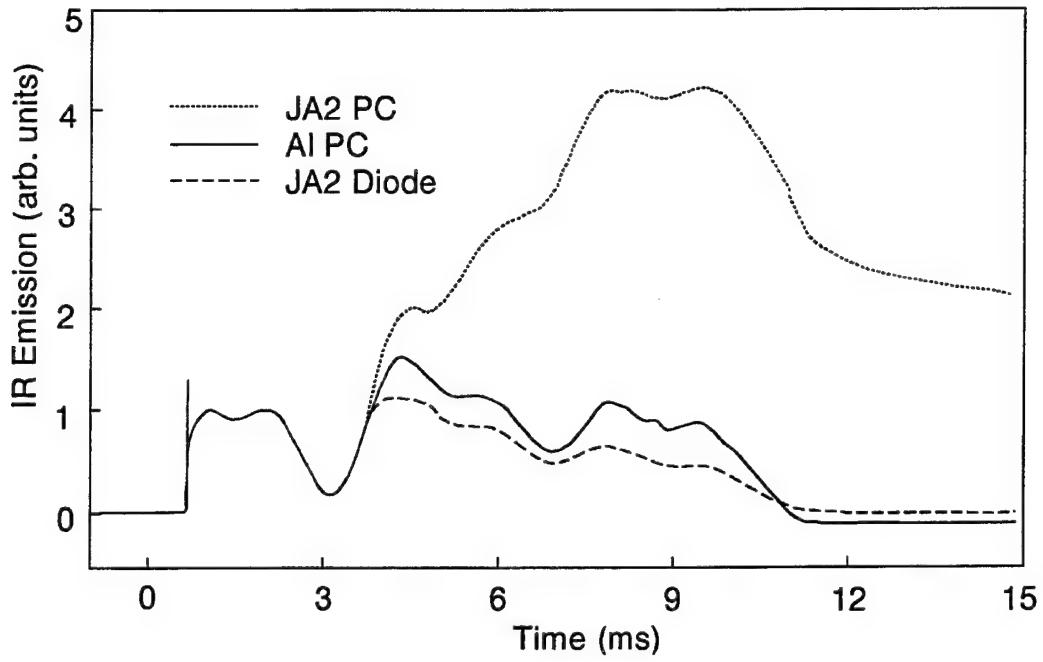

Figure 11. IR Emission at 4.49 μm From JA2 Propellant and an Aluminum Disk in Air During a 10-ms Laser Pulse. Intensities Are Relative to Values at 1 ms. As in Figure 10, the Emission Near 4 ms Is Attributed to Initiation of Exothermic Reactions.

Table 1. Effect of Flux Density on Flamespreading

Identification No.	Diameter (mm)	Power (kw)	Flux ($kcal/cm^2$-s)	Observation
66	3.5	1.63	4.1	f
67	3.2	1.63	4.9	c
68	3.5	1.63	4.1	c
69	5.5	1.63	1.5	f
70	5.5	1.32	1.3	f
71	5.5	1.32	1.3	f

Note: f = flamespreading and c = crater formation (ablation).

4. Discussion

For the range of surface temperatures (400–800 K) in Figure 2, the ratio of the (Ahrrenius) propellant reaction rates (N to NC) is between 0.01–25. The "inert" delay calculations for $n = 250\ cm^{-1}$ (Figure 1) indicate that differences in kinetic parameters on adiabatic model predictions are only noticeable at low flux values. At high values, the delays are determined primarily by inert heating. The calculated delays are sensitive to values of n. Considering the scatter in the data, the model predictions are in reasonable agreement with measurements and, as seen in Figure 3, are almost as good as the agreement obtained using the surface-temperature criterion determined from the experimental data.

The calculations in Figures 6 and 7 indicate that, with the GNL-10 laser ($E_o = 13.5$ J, 10-ms pulse), values of n greater than 15 cm^{-1} are required for T_b to exceed the average T_{crit} (561 K) and values of n greater than 5 cm^{-1} are required for the inert delay (t_{in}) to be less than t_{max} (9 ms).

Most solid materials can be made to emit and ablate under sufficiently high-power/flux irradiation. This makes it difficult to determine if the initial emission and pressurization with energetic solids is the result of initiation of exothermic or ablation (endothermic) reactions. The pressure, photomultiplier (visible emission), and photoconductive detector (IR emission) signals shown in Figures 8–11 indicate there are some differences in the response of JA2 and inert solids to Nd:glass laser irradiation. If these differences occur during the pulse and are independent of ambient gas, then the condensed-phase adiabatic model may be able to predict the delays in propellant response and, perhaps, initial pressurization rates under these conditions. Differences in the pressure response of JA2 and inert propellants can be seen in Figure 8. Shortly after deradiation, the inert pressure goes through a maximum followed by monotonic decrease. With JA2, pressure increases monotonically and maxima occur at later times. The JA2 pressure signals indicate that the initial pressurization rates depend on flux density but not on ambient gas composition.

In these experiments with JA2, the presence of O_2 leads to flamespreading. (The pressure signals suggest that, at high flux density, flamespreading occurs during irradiation but at low flux density

is delayed [~75 ms] after deradiation.) For the same flux density, there is a difference between JA2 and inert-propellant initial pressurization rates, but, due to the filtering, the differences in delays cannot be determined.

The photomultiplier signals in Figure 9 suggest that the origins of inert and JA2 visible emission during the pulse are different. At the end of the laser pulse, the decay in inert-propellant emission is delayed, but, once started, the emission is rapidly extinguished. This is suggestive of thermal emission from a cooling body. With JA2, the delay is shorter but the emission is not rapidly extinguished and remains at a low level. This low-level emission may be the result of exothermic reactions initiated during the pulse. The subsequent increase in emission in air is due to initiation of flamespreading.

Evidence of differences in the response of JA2 and inert propellants during the laser pulse can be obtained from the photoconductive detector records. In Figures 10 and 11, gasification/ablation of the JA2 and inert-propellant surfaces occurs during the pulse but the aluminum surface is unreactive. The (relative) signals show that, for times less than 4 ms, the IR emission at 4.49 μm (from inert and JA2 propellant and aluminum) follow the laser intensity fluctuations recorded by the diode detector. At later times, deviations occur in the JA2 signals. The initial part of these signals may be due to thermal emission. The deviations observed with JA2 at later times do not appear to be due to the ablation process/products and may be due to initiation of exothermic reactions at times greater than 4 ms. For the flux intensity of these experiments (2.0-2.7 kcal/cm^2-s), the adiabatic model prediction for the inert delay for n = 15 cm^{-1} (experimental JA2 value) is about 6–7 ms. The agreement seems reasonable, considering that the assumptions made in the model, but it may be fortuitous. The effect of the laser intensity fluctuations needs to be determined before meaningful comparisons can be made.

5. Conclusions

The initial experimental results with Nd:glass lasers at flux densities between 1–5 kcal/cm^2-s indicate that exothermic reactions at the JA2 surface are initiated during the laser pulse. These

reactions affect the emission response, but, at present, it cannot be determined if they affect the pressure response. The pressure (Figure 8), emission (Figure 9), and IR (Figure 10) responses appear to be independent of ambient gas composition during the laser pulse. This suggests that condensed-phase models can be used to predict emission delays. Calculations of the inert surface temperature indicate that critical temperatures for emission determined from JA2 experiments using a CO_2 laser will be exceeded during the laser pulse only if absorption coefficients exceed the experimentally derived value (15 cm^{-1}). The calculated delays using a condensed-phase model are in reasonable agreement with measured IR emission signals. The calculations are sensitive to values used for optical properties.

The data in Table 1 suggest that there may be an upper limit on flux density values for flamespreading to occur. Exceeding the limit leads only to ablation. Understanding the ablation process is important in predicting initial pressurization rates and delays. At present, it is not clear if and how the gasification/ablation process for energetic and inert solids differs. Experiments with inert and energetic solids that have similar optical, as well as thermophysical properties, will be needed to answer that question.

The use of a radiant ignition model to predict the initial pressurization (delays and rates) due to a high-power pulsed laser remains a challenge.

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13. ABSTRACT *(Maximum 200 words)*

A study of JA2 ignition using high-power (3 kW) pulsed Nd:glass lasers at low loading densities was undertaken to help construct models for predicting initial pressurization in bombs at high loading densities. Comparisons were made with results of experiments using a low-power (0.1 kW) continuous wave (cw) CO_2 laser. Differences in wavelength dependence of optical properties and time dependence of the laser pulse were considered. Measured infrared (IR) emission delays agreed with the calculated "reaction" delays using a condensed-phase model. The calculations are sensitive to values used for optical properties. Experiments in which laser energy and beam diameter were varied indicate that there is an upper limit on flux density values for flamespreading to occur. Exceeding the limit leads only to ablation. An understanding of the ablation process due to high-power irradiation of energetic solids is needed to predict the pressure response of solid propellant to high-power lasers.

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